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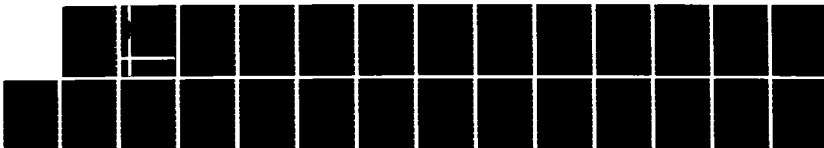
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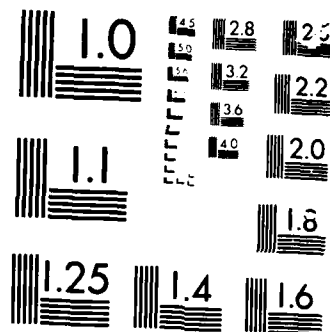
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THE INFLUENCE OF WELDING FLUX ON THE
PYROMETALLURGICAL, PHYSICAL AND MECHANICAL BEHAVIOR OF WELD METAL

FINAL REPORT

DAVID L. OLSON

May 15, 1986

U.S. ARMY RESEARCH OFFICE

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Aluminum weld metal cracking has been characterized for both binary and commercial aluminum alloys. A new model for the prediction of the susceptible and extent of hot cracking has been developed.

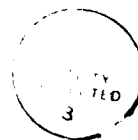
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Abstract

The pyrochemical behavior of submerged arc welding flux was investigated. The nature of the pyrochemical reactions are described. Mechanistic models for the pyrochemical behavior are suggested. The submerged arc welding was performed on both low carbon and high strength low alloy steels. Both thermodynamic and kinetic models are discussed.

Ferrous weld metal phase transformations were investigated. The role of weld metal oxygen, weld metal inclusion and strain on the formation of acicular ferrite was characterized, analyzed and modeled. Advanced high purity, homogeneous multi-component welding fluxes were made using the sol gel process for making glass. These sol gel fluxes were used to make flux cored wires. The sol gel filled flux cored wires were used to weld microalloyed HSLA steels and were found to have excellent arc stability and low moisture content. Sol gel fluxes offer the consistency necessary for welding systems which will use in situ sensing and microprocessor control. A capacitance technique has been developed for moisture content measurements of the coating on covered electrodes.

Aluminum weld metal cracking has been characterized for both binary and commercial aluminum alloys. A new model for the prediction of the susceptibility and extent of hot cracking has been developed.

1.0 Pyrochemistry of Submerged Arc Flux Welding of Ferrous Alloys

Welding fluxes have been designed to satisfy a multiplicity of requirements such as arc stability, weld metal protection, weld metal deoxidation and alloying, slag detachability, etc. These requirements would be better fulfilled with further understanding of the many chemical reactions occurring simultaneously during welding. By performing experiments in which

only a small number of parameters are varied in a well defined manner, understanding of the kinetic and thermodynamic factors which influence the composition of the weld metal and the quality of the weld was obtained (1-3). The thermodynamics and kinetics of the chemical and phase reactions which may be operative during welding were analyzed to place limits on the range of possible reactions and processes which might occur in the complex range of materials, densities, temperatures, and states which lie in the relatively small distance between the filler metal feed and the metal to be welded.

Despite suggestions to the contrary, it is unlikely that equilibrium could be attained during welding even at the high temperatures. Despite the expected departures from equilibrium, one may utilize equilibrium considerations to place constraints on the chemical reactions and mechanisms involved in welding. A common approach is to assume that a state of thermodynamic equilibrium is attained locally, on the basis that the high temperatures and high surface-to-volume ratio counteract the short time available for reactions to be completed.

Measurements (2) have been made of the compositional changes of fluxes and weld metal during submerged arc flux welding of low carbon steel with a series of synthetic MnO-FeO-SiO_2 model fluxes containing 40 wt. pct. SiO_2 and different ratios of MnO to FeO. Simple and plausible mechanisms which could control the weld metal content of manganese, carbon, silicon, oxygen, phosphorus and sulfur in our experiments have been deduced (2). These mechanisms involve a deduction of the thermodynamic driving forces and consideration of the kinetic factors which might be operative in these nonequilibrium systems. One unique aspect of this picture is that transfer into or out of the metal phase is directly controlled by interfacial and not bulk activities of constituents (such as FeO). The concentrations of carbon,

phosphorus, sulfur and possibly silicon tend to be larger in the molten metal than in the base plate.

The effect of CaF_2 , CaO , and FeO additions to the manganese-silicate welding flux system on AISI 4340 steel weld metal was also investigated (4,5). A comparison of the effects of these flux additions on AISI 4340, and 1020 steel welds was made in an effort to understand the effect of alloying elements on weld metal chemistry. Data are reported as a resource for future analytical and comparative purposes.

This investigation has not considered some of the complexities inherent in such a complex multicomponent multiphase system (3). For example, although ternary interaction terms between the solutes in the metal are usually small, these could alter the thermodynamic properties of some of the solutes significantly. In addition, diffusion coefficients in ternary silicate systems, such as the slag, could be complicated by ternary effects. Also, the concentration of silica at the interface could be very different from the nominal value of 40 weight percent. Ultimately, one must consider such complexities to obtain a more complete understanding of the phenomena and pyrometallurgical reactions involved in welding. A further complication is provided by the high voltages and currents employed. These could readily generate chemically activated metastable materials by a variety of possible electrochemical reactions. For example, metastable metal atoms or clusters would have a much higher volatility than stable metals.

Perhaps the most important conclusion is that it should be possible to control changes in weld metal composition. For example, the manganese content can be controlled by fixing the ratio of the activities of MnO to FeO to be the same as that at the neutral point (where ΔMn is equal to zero). Thus, the detailed knowledge of the chemical reactions occurring during welding can

be used to control compositions of weld metal and improve the quality of welds (3).

The kinetic factors important for submerged arc flux welding have been briefly discussed (3). Other factors can influence the chemistry of weld metal. For example, vaporization of FeO and MnO from the flux to the ambient atmosphere at high temperatures is larger than the vaporization of SiO_2 . The consequent changes in slag chemistry could influence the chemistry of the weld metal. In addition, there are many metastable species in the arc because of the tendency of electrons to ionize atoms. Currently, there is insufficient information on these factors for useful analyses. Knowledge of the diffusion coefficients of different components in the two contacting phases are important for determining the relative kinetics and are also limited.

It was deduced that the activity of FeO at a slag-metal interface and the pickup of oxygen by weld metal will be determined by the most oxidizing component in the slag (2). Even when no FeO was present, the MnO will react with iron will lead to a significant activity of FeO at the interface. This method of analysis of the mechanism for manganese and oxygen transfer into or out of the metal phase should prove to be a unique aid in the reactions considered as well as in a number of analogous reactions. Slag compositions can be changed to minimize the interfacial FeO concentration even while maintaining a ratio of $a_{\text{MnO}}/a_{\text{FeO}}$ which will keep the manganese content of the metal constant. For example, one can decrease the contents and activities of MnO and FeO by partly replacing them with very stable oxides, such as MgO.

One significant experimental result is the transfer of sulfur, phosphorus and carbon from the solid base plate into the weld pool. It is important to study welding conditions which influence transfer of these and other elements

into the molten weld pool and their segregation during crystallization. Such compositional changes influence crystallization characteristics of the weld pool and the subsequent performance of the weld. A full understanding of this elemental transfer and segregation is important in improving the quality of submerged arc welds (3).

2.0 Role of Weld Metal Oxygen in Weld Metal Phase Transformation

High toughness in low alloy steel weldments can be related to the amount of weld metal acicular ferrite. Twenty eight reagent grade fused fluxes from the $\text{CaF}_2\text{-CaO-SiO}_2$ system were used to produce bead-on-plate and double-V-groove submerged arc welds on a quenched-and-tempered niobium HSLA steel to investigate the role of weld metal oxygen on the formation of acicular ferrite (6-9). An E70S3 welding wire was used with two different heat inputs - namely, 1.9 and 3.3 kJ/mm (48.3 and 83.8 kJ/in.). A niobium microalloyed steel was selected because of its fine grained microstructure, high yield strength, and high toughness at low temperatures. Fluxes from the $\text{CaF}_2\text{-CaO-SiO}_2$ system were selected because of their low oxygen potential, and their ability to produce low oxygen (80-450 ppm) welds. Quantitative metallography and chemical analysis were performed on the welds. The chemical behavior of this flux system has been characterized with respect to manganese, oxygen, silicon, niobium, and sulfur.

The lower heat input welds showed a predominately fine microstructure of acicular ferrite. At high oxygen content, a higher percentage of grain boundary ferrite (ferrite veining) was observed. By reducing the oxygen in the weld metal, the amount of acicular ferrite was increased. With further reduction of weld metal oxygen, the main microstructural feature, instead of acicular ferrite, became bainite. Using higher heat input, the weld metal microstructure transition with oxygen level was not so clear.

In spite of the essentially similar optical microstructure and similar chemical composition (other than oxygen), the mechanical properties of the various welds were observed to be very different. Toughness data (upper shelf energy and transition temperature) were found to correlate with weld metal oxygen content. The upper shelf energy decreased with increasing oxygen level in the weld metal.

The results of this weld metal oxygen investigation led to the following conclusions:

1. The $\text{CaF}_2\text{-CaO-SiO}_2$ flux system is found to produce good quality niobium microalloyed HSLA steel weldments with very low oxygen content (100-460 ppm).
2. Manganese and niobium showed negative delta values during welding, indicating a loss from the weld pool to the slag.
3. There is a region of zero delta silicon where weld pool chemistry can be easily controlled.
4. The negative delta sulfur values indicated that the $\text{CaF}_2\text{-CaO-SiO}_2$ flux system was effective in sulfur control.
5. The optimum weld metal oxygen content for this flux system ranged from 200-300 ppm, corresponding to a microstructure of approximately 90 percent acicular ferrite.

6. The refinement of the weld metal microstructure due to oxygen decrease resulted in an improvement of the weld metal toughness.
7. High weld metal oxygen welds showed high inclusion contents, but the inclusions were observed to be finer in size.
8. In the lower oxygen content welds, a larger strain was found to associate with the upper shelf microstructure of ductile dimples leading to a higher energy absorbed at fracture.
9. The lower shelf failure was characterized by cleavage fracture with small regions of ductility between the flat fracture facets which account for the energy difference between high oxygen and low oxygen welds.

3.0 Role of Inclusions in Weld Metal Phase Transformations

Since non-metallic inclusions are known to influence the formation of acicular ferrite, the chemical and physical features of inclusions that affect the weld metal ferrite phase transformations were investigated (10,11). Submerged arc welds have been made with five selected wire compositions (Fe-Mn-Si type wires), each of which was selected to introduce various oxide forming (Al, Zr, Ti, Mo, B) additions into the weld pool. Welds were made on niobium microalloyed steels with a $\text{CaF}_2\text{-CaO-SiO}_2$ reagent grade welding flux. Chemical and metallographic analyses were performed on weld metal compositions and microstructures. The composition, morphology, size and spatial distributions of inclusions are being determined as a function of these weld pool additions using SEM-EDX analysis. Transmission electron

microscopy was performed on inclusions extracted from weld metal using a carbon extraction replica technique. The composition variations in the matrix adjacent to the inclusion is being determined. Some evidence of inclusions nucleating acicular ferrite laths has been observed. With the exception of few, most inclusions were found to be spherical, and etch pits with definite facets could be seen around these inclusions. The carbon extraction replica and thin foil techniques were used to characterize the inclusions. This effort is essential in the understanding of the nucleation mechanism of acicular ferrite.

Inclusion size distribution was found to be a very important factor in achieving a desired weld metal microstructure (10,11). Inclusions may locate at the grain boundaries or within the austenite grains. Depending on the size distribution, the partition of inclusions to the boundary and to the interior of grains may be different. Small particles more often associate with the grain boundaries, limiting the grain size and produce coarse grain boundary allotriomorphic ferrite. Large particles do not pin the grain boundaries as well and promote the formation of intragranular acicular ferrite, bainite, etc. An optimum size distribution was found to depend on the types of inclusions and is closely related to the weld pool deoxidation practice.

The free energy associated with inclusion/matrix differential thermal contraction was found to be insufficient when compared with the austenite - ferrite transformation volume free energy change. Thus, strain energy contribution due to thermal contraction at cooling could not be used to explain the inclusion size distribution effects observed, nor could it be used to correlate with acicular ferrite formation.

To improve the ability of predicting weld metal phase constituents, Avrami equation for surface nucleation was modified to describe the

transformation of an austenite grain into grain boundary allotriomorphic ferrite, combining the effects of cooling rate and prior austenite grain size (10,11).

4.0 Influence of Strain on Weld Metal Phase Transformation

The role of welding flux on maintaining low weld metal oxygen content and the influence of weld metal oxygen and inclusions on the formation of weld metal acicular ferrite (the critical microstructure to achieve high weld metal toughness) has been carefully characterized and modeled. Another area that has been investigated is the effect of strain energy on the phase transformation behavior of low carbon steel weld metal. Various strain conditions may change the size, fraction, shape, or distribution of acicular ferrite in the weld metal. This weld metal phase transformation research effort was designed to determine the contributions that macro- and micro-strain energy situations have on the formation of acicular ferrite. The macro-strain energy conditions occur across the weld bead due to shrinkage and thermal expansion and micro-strain energy conditions are those resulting from the thermal expansion or contraction mismatch between the weld metal austenite and the inclusion. Since inclusions have been identified as the heterogeneous nucleation site for acicular ferrite, it is anticipated that any strain energy in the austenite adjacent to the inclusion is going to have a direct influence on the amount of undercooling for the ferrite transformation and on the number density and size of acicular ferrite formed. If strain energy effect exists and is understood, then modifications can be made to either the consumable or to the joint design to optimize mechanical properties.

Two main concerns were addressed: First, does the differential thermal contraction between austenite and inclusions modify phase transformation

behavior by creating localized strain fields? This has been modeled using a ball-in-hole analogy. Secondly, does the presence of residual stress in a weld modify the decomposition of austenite? Gleeble Thermal-Mechanical Testing and electron microscopy have been used to perform appropriate experiments to investigate the role of residual stress. This investigation has produced direct evidence that increasing tensile or compressive strain will increase the amount of weld metal acicular ferrite.

5.0 Sol-Gel Welding Flux

New methods to make high purity compositional controlled, advanced welding fluxes have been investigated. The sol gel method of making glass and ceramic materials was explored as a method to make advanced welding fluxes (12,13). The sol-gel welding flux development effort started with a thorough literature search and selection of proper chemicals and safe chemical processing. High quality submerged arc welds were first produced using aluminum silicate and calcium silicate sol gel glass fluxes. X-ray spectrochemical and structural analyses have found that these fluxes are amorphous, high purity silicates. Submerged arc welds which have been made with these initial fluxes were analyzed.

The sol gel process was then used to produce high purity, homogeneous $\text{CaO-TiO}_2\text{-SiO}_2\text{-1\%Na}_2\text{O}$ glass welding fluxes for flux cored wire. Ten high purity sol gel fluxes of different compositions have been prepared in sufficient quantities to produce 25 lb batches of flux cored welding wire. Ten flux cored wires based on these different sol gel fluxes, were made with the assistance of Alloy Rods Corporation of Hanover, Pennsylvania.

Flux cored arc welding with these experimental wires was performed on niobium microalloyed HSLA steel. The physical behavior of these sol gel

welding consumables and the resulting weld metal was thoroughly characterized. These $\text{CaO-TiO}_2\text{-SiO}_2\text{-1\%Na}_2\text{O}$ sol gel welding fluxes also offered the purity and consistency necessary for future high production welding processes which will use microprocessor control.

The moisture pick up of these sol gel fluxes was found to be insignificant by a four week moisture test. Welds were made with three different heat inputs (20, 40, and 60 KJ/in.) using the sol gel flux cored wires with an argon cover gas. Arc stability was excellent which suggests that sol gel consumables have promise as a consistent consumable material for automatic welding processes with feedback controls. Welds made by these sol gel fluxes had hydrogen contents of approximately 6 ml/100 grams of weld metal. With optimization of the sol-gel process, the hydrogen contents can be kept below the 5 ml/100 grams of weld metal which is the required limit for high strength steel weldments. The weld metal microstructure was also characterized as a function of flux composition (12,13).

6.0 Moisture Content Meter for Covered Electrodes

A capacitance method to measure moisture content on covered electrodes has been developed (14,15). Direct correlation between moisture content and capacitance for three low hydrogen type electrodes (E7018, E8018, and E10018) has been reported. Similar correlations between hydrogen content of weld deposit and capacitance has been determined. This technique should be very useful in monitoring the moisture condition of electrodes in service and allow for better hydrogen control.

7.0 Gas Metal Arc Welding Metallurgy

The effect of variations in cover gas oxygen activity (Argon- O_2 or Argon- CO_2) and heat input (cooling rate) of the Gas Metal Arc welding process on the microstructure and properties of niobium microalloyed weld metal was investigated (16,17). This investigation, which is based on the oxygen activity of shielding gas, will determine the influence of oxygen on the weld metal composition and microstructure independent of a flux. These results are being compared with our submerged arc welding results to determine a better understanding of the behavior of welding fluxes. This investigation has demonstrated that the gas metal arc welding process can achieve the optimum weld metal microstructure and properties in niobium microalloyed steels with the proper selection of welding wire, shielding gas and heat input.

The nucleation and growth phenomena that characterize the transformation of austenite in HSLA steels are influenced by both cooling rate and composition. An important part of the composition influence is due to the oxide particle nucleants which form during the welding process. The composition of weld wire and shield gas in gas metal arc welds of HSLA steels determine the oxide particle formation; consequently, the weld wire and shield gas combination is more important than the choice of either wire or gas alone. A plot of inclusion-forming elemental additions (effective oxygen) versus hardenability additions (effective manganese) was found to be useful to predict GMA weld metal microstructure. Also, the weld metal composition was found to be influenced by chemical reactions in the weld pool, not just at the end of the electrode as has been suggested in the literature.

As a matter of practical interest there is only a limited number of parameters under the control of the welding engineer. This work showed that the preferred microstructure is a function of the heat input and cover gas for

each of the welding wires used. It is apparent that each wire has an optimum region. Thus, it is important that the choice of welding wire for a particular application be coupled with the choice of shield gas. For example, it can be seen from this work that with the argon- O_2 cover gas, a greater range in heat input can give acceptable welds than the use of an argon- CO_2 cover gas.

8.0 Aluminum Weld Metal Cracking

Difficulties with hot tearing are commonly encountered when welding high strength aluminum alloys such as alloy 2024 or 7075. Thus, in developing new high strength aluminum-lithium alloys to replace alloys 2024 and 7075, their susceptibility to hot tearing needs to be investigated to determine if these new alloys can be welded.

The weldability of high purity aluminum-lithium binary alloys has been investigated using the varestraint test (18). Alloys were prepared by mixing molten aluminum with molten lithium in an inert atmosphere. After mixing, the alloys were chill-cast into the form of rectangular ingots suitable for use in the varestraint test. GTA varestraint tests were run on cast ingots of aluminum-lithium binary alloys ranging from 1 to 5 w/o Li. Test results revealed the typical peak susceptibility behavior of total crack length versus composition common for aluminum alloys. At maximum susceptibility, however, aluminum-lithium alloys were found to behave significantly better than the weldable aluminum alloy 5083, tested under identical conditions. Results have shown that maximum susceptibility to hot tearing occurs at approximately 2.6 weight percent lithium. It is a significant engineering finding that high purity aluminum-lithium alloys are readily weldable. Both weld pool geometry and weld metal grain structure varied with increasing lithium concentration,

apparently by a sharp drop in thermal conductivity. This work was reported at the 2nd International Conference on Aluminum-Lithium Alloys (18).

A hot cracking model was developed to determine the hot cracking susceptibility of aluminum weldments. The model identifies the impurity (binary or ternary) eutectics as the primary factor in the formation of these cracks. This model, which also incorporates many of verified features of previous models, was tested. A test matrix involving numerous aluminum alloy compositions with known levels of impurities was used in this investigation. The Al-Li alloys will be given special attention in order to understand their excellent weldability. This investigation used a variety of experimental techniques which included varestraint testing, Gleeble simulation of thermal cycles, harpoon thermal analysis of the weld metal, bulk and fracture surface chemical analysis, and optical and electron microscopy.

This model also relates maximum crack length, X_{\max} , to the thermal gradient (at the weld pool interface), the extent of the liquid-to-solid transition zone, and the difference between binary and impurity eutectic temperatures (ΔT):

$$X_{\max} = (m_L \Delta C + \Delta T) / (dT/dx)$$

where ΔC is the difference between eutectic and nominal compositions and m_L is the slope of liquidus line. It is assumed that the weld pool boundary is at the binary eutectic temperature and the outer most tip of a crack is at the impurity eutectic temperature. Experimentation has shown some agreement for this proposed model in the Al-Cu, Al-Li, Al-Mg, and Al-Si alloy systems (19-21) with impurity additions intentionally made to see if the respective ΔT affects X_{\max} in the manner predicted. This investigation has identified specific eutectic forming elements causing cracking in aluminum alloys.

The high strain rate imposed during varestraint testing may be responsible for extended crack formation due to mechanical energy dropped in

the crack susceptible region of the test specimen. To examine this possibility, high temperature/strain rate tensile tests was run utilizing a Gleeble thermal-mechanical testing system. Test specimens were machined directly from the weld metal of each of the four different binary alloy systems. A direct correlation between ductility and susceptibility to vareststraint cracking was not found.

Another investigation has been studied on the influence of grain refiners (Zr, Ti, B and Ta) on the cracking susceptibility of aluminum alloy weld metal. The grain refining ability of titanium and zirconium additions has been weld characterized for 1000 aluminum weld metal (22).

9.0 Other Research Activities

A literature review of the mechanism of weld metal pore formation is near completion (23) and will be published in a book belonging to a Review Series on Manufacturing Processes. A review article on "Selecting Arc Welding Processes" was published in Chemical Engineering (24). To ASM handbook chapters were written (25,26) and two major reports (27,28). Also two articles were published based on results of a prior ARO contract (29,30).

10.0 Scientific Personnel Supported by this Project and Degrees Awarded During This Contract Period

Employment

C.E. Cross, Ph.D. (June, 1986)	IMAC Research and Consulting
C.S. Liu, Ph.D. (July, 1985)	Assistant Professor, Pennsylvania University
C.B. Dallam, Ph.D. (July, 1986)	To finish soon
P.S. Dunn, M.S. (Dec., 1985)	Los Alamos National Laboratory
D.E. Bunnell, M.S. (Dec., 1984)	Rockwell International (Rocky Flats Plant)
P.A. Burke, M.S. (Sept., 1985)	Spectra-Physics Corporation (Laser Systems Div.)
R.E. Francis, M.S. (May, 1984)	Ball Brothers Corporation

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